ANOMALIES IN THE ASSESSMENT OF COKING POWER IN LABORATORY TESTS

Michael Pretor

ABSTRACT

Laboratory carbonisation tests such as the Plastometer (Gieseler, Brabender, Sapozhnikov) Dilatometer (Audibert - Arnu, Ruhr) and Swelling Index are designed to reveal the rheological behaviour of coking coals under prescribed "experimental condition". Tests such as the Gray-King Coke Type, Roga Index and Caking Index (previously British Standard now Indian Standard) are used to measure the caking (agglutinating) capacity of coking coals (also used under specified experimental conditions).

All of these tests have become more or less reliable tools of the industrial technology in the "old world" endowed with coking coals of the Carboniferous. Geographical popularity of certain tests have been influenced by specific features of the indigenous coal until the event of global trade in raw materials and industrial "know-how".

Application of tests such as the Dilatometer or Gray-King Coke Type to coals of a different Genealogy invariably under rates the true coking capacity. In the introduction process between seller and buyer success will not be easy with mediocre results.

The purpose of this paper is to re-examine the "empirical" nature of some of these tests and highlight the potential for misleading conclusions.

The examples discussed all pertain to a single seam coal deposit in the Southern Coal Fields of New South Wales, Australia, which is of great commercial significance to the local Steel Industry as well as to a number of Exporters.

DISCUSSION

A number of tests have been developed to assess the behaviour of coals in the coking process, some of which have found acceptance only in countries where they were developed, while others are being used in most parts of the world. Occasionally different values are reported for a sample of the same coal and the progress of introducing new coal to a prospective buyer is hindered. More often the absolute differences are not the main concern, but the interpretation put on the utilisation consequences of the "other than expected result".

No single test by itself is adequate to determine coking capacity; a suite of tests are necessary to assess various aspects of the coal when evaluated for coke making.

Generally, six sets of Laboratory indices are considered adequate in any selection consideration, and these are;

1. Crucible Swelling Index
2. Gray-King Coke Type or Roga Index (Indian Caking Index)
3. Plastometric properties, most commonly Gieseler, but in certain countries Saphozhnikov or Brabender.
4. Dilatometer characteristics by Audibert - Arnu or Ruhr (also British Standard).
5. Maceral Composition of the Coal
6. Reflectance of Vitrinite.

Because of the highly empirical nature of the first four tests and the subjective (individualistic) nature of the fifth and sixth, some experimental features of each are briefly reviewed.

1. Crucible Swelling Number (Index)

By far the most popular test which is indicative of coking potential. It is a simple test and very rapid. However, test result is highly sensitive to the size distribution of the test portion, rate of heating and final temperature. Laboratories equipped with high speed ring grinders for the preparation of coke samples might prepare coal samples in the same, and obtain values several units lower that the true result. In exploration work such practice can be lead to the abandonment of a prospective coking coal deposit.

A very serious mis-interpretation of a lower than expected Swelling Number is the assumption that the coal sample has become oxidised and thus, lost it's coking properties. Although Crucible Swelling Number is used as a parameter in Classification Systems, for the abovementioned reasons it is not entirely dependable for assessing coking properties.

Euro-American coking coals generally possess Swelling numbers of 7 or higher, by contrast in Australian coke making coals with Swelling Numbers below 7 (to as low as 4) are the norm.

Table 1 and Graph 1 illustrate the relationship between particle size distribution of test portion and Crucible Swelling Number.

2. Gray-King Coke Type/Roga Test Caking Index

Both tests are more complex and take longer to perform than the Swelling Index and are also used as classification parameters. The results from both tests are believed to be more reliable because of greater control over the heating conditions.

However, size distribution of the test portion needs to be controlled for the previously mentioned reasons and testing is to be conducted on the same day as the final stage preparation. The GKCT test requires the addition of measured amounts of Electrode Carbon for strongly swelling coals to maintain the original volume of the coal and carbon mixture. Values from A to G and then G_1 to G_{10} according to the mass of electrode carbon used.

Evaluation of the GKCT results is highly subjective in terms of strength and hardness as judged by the sound produced when tapped on a hard wooden surface.

Because of the difficulties in the accurate identification and measurement of "inertinite" in Australian coking coals and given the generally poor GKCT values for the Bulli Seam Coal, experiments were conducted to make a more objective assessment of the reliability of this test for coking coal classification.

Two coal samples were carbonised in the prescribed manner with 10, 20, 30 and 40% electrode carbon. The resultant GKCT coke were visually evaluated and then sliced into section suitable for Tensile Strength measurement (Brazilian compression test) and bulk density determination.

As illustrated by Table 2 and Graph 2, both coal samples produce stronger cokes with the addition of electrode carbon. In other words, although categorised as "inert-rich" each in fact needs additional inerts for maximum coke strength. It is also significant (and not unexpected) that maximum coke strength corresponds to the highest bulk density (minimum porosity). More about this under Macerals and Microscopy.

3. Plastic Properties

As measured by the Gieseler Plastometer are perhaps the most often discussed for reasons of instrumental differences and suitability for the detection of oxidation. KCC's experience with the measurement of plasticity (maximum fluidity) has been extensive on account of the large sealed stockpiles that the company held in previous years. The long-term preservation of coking properties is of utmost importance to all coking coal producers.

In the course of building up long-term stockpiles (approximately 1/2 million tonne each) preventive and control measures were instituted to monitor coking properties. These included the periodic (3 months) sampling and testing of samples from pre-determined positions, over a period of 18 months. The early results were highly variable in terms of Gieseler fluidity in comparison with Crucible Swelling number for example, suggesting either oxidation or experimental error.

On closer examination of the procedure it was recognised that the periodic re-sampling of the stockpile in close proximity of the previous sample positions was distorting the results (i.e.; exposure of the localised area in the stockpile was responsible for the erratic results). Therefore a modified, more intensive - but further spaced sampling programme was conducted which gave statistically acceptable results.

Table 4 and Graph 4 illustrate the results.

It might be of interest to note the technique of sampling large (compacted) stockpiles, as shown in the slides. Dry ice (solid CO_2) was mixed with the samples immediately and stored under refrigeration until the time of testing. To validate the effectiveness of this procedure two sets of samples were processed and stored under ambient conditions. Results are shown in Table 5.

4. Dilatation Test

Dilatometer determined indices have been the basis of coal blending in Europe for many years and gained popularity elsewhere. The work of Scientists in West Germany, Holland, England and more recently Japan, are well known and yet this day the grind of the test sample is such that certain coals will be under estimated.

This test is also highly regarded by certain Research Centres for monitoring deterioration in coking properties.

IN the course of the earlier mentioned stockpile surveillance program, bulk samples of freshly washed and also stockpile coals were submitted for evaluation including the Dilatometer derived "oxidation resistance" prediction.

Figure 5 illustrates the experimentally derived curve as well as the calculated curve projecting the ambient temperature, natural (unprotected) storage stability.

It should be noted that a full range of analyses, including Proximate, Ultimate analysis , Specific Energy, Carbonisation tests (Swell, Gieseler and Dilatation) were carried out for correlation purposes. Pilot-scale carbonisation was also carried out on both stockpiled and freshly-washed coal.

The conclusions reached was that "there is no difference between the fresh and stockpiled coal", although there were some differences in the Swelling number and Dilatation.

The most remarkable observation in this evaluation was that the "coals with approximately 25% dilatation can replace coals with 25,77 and in part even 127% dilatation, without this having any negative implications for the coke strength", in blends with good coking properties.

Since that time all the 2.4 million tonne of stockpiled coal had been successfully coked by a number of Cokeworks for Steelmakers in different countries.

5. Maceral Composition

Petrographic analysis, by counting the amount of microscopically identifiable constituents and grouping into "reactive" and "inert" categories is used to determine whether the coal has sufficient reactives to provide the bonding to make strong coke.

Among the numerous methods of predicting the coking quality of a coal the Schapiro-Gray system has acquired a wide following, both in it's original form or with some modifications.

Similarly to the carbonisation tests discussed, the method has been modelled on Carboniferous coals and consistently underestimates the coke stabilities of inertinite-rich coking coals of the Southern Hemisphere. The main reasons for the erroneous predictions are an underestimation of the contribution to coke making of the maceral group inertinite.

The following comments are based on the experimental work of Professor Diessel (University of Newcastle, Australia), using a number of coals from different countries, including the Bulli Seam from New South Wales.

the difficulty with the petrographic method is that some macerals are not readily assigned to either the reactive or inert groups.

Ring Analyses organised by the International Committee for the Coal Petrography produced erroneous coke stability prediction when Southern Hemisphere coals were involved. This has been the subject of Diessel's investigations and results pertaining to the Bulli Seam are shown in Figure 6 and 7.

It has been found that a suitable indicator of reactivity of intertinite is the degree of anisotropy (bi-reflecltance) after coking. Inertinite with a pre-carbonisation reflectance (PCR) of more than 1.8% $R_o$max remains isotropic during carbonisation.

Inertinite with less than 1.8% PCR becomes highly anisotropic on coking and also displays signs of sufficient melting to ensure complete textural integration and bonding. The experimentally determined amount of reactive inertinite in this study was 43% which is considerably different from the traditional 1/3 of semifusinite allowed in the Schapiro-Gray method.

Furthermore, even the unfused inertinite is not altogether detrimental because the presence of fine-grained inertinite which can be completely wetted by the molten coal during carbonisation, increases the wall thickness between degassing pores and thus adds strength to the coke. Such fine-grained inertinite is recorded as "micrinite" and "inertodetrinite" in maceral analysis, the maximum particle sizes of which are 2μm and 30μm respectively.

In order to predict realistic coke stability indices for the Bulli Seam the Schapiro-Gray method can be used provided that a change is made to the method of determining the proportion of reactive components, i.e.; "reactives" consist of:-

(a) all vitrinite
(b) all exinite
(c) 43% of all inertinite, and
(d) 30% of all inertodetrinite plus micrinite

Coke stabilities calculated by using these parameters and conventional method are in Table 5, together with actual stabilities determined on 230kg Pilot-oven cokes.

Petrographers not fully versed with the subtleties of certain inertinite-rich coals could not undertake such modifications to their procedures, hence the systematic underestimates of ASTM stability of the Bulli Seam.

6. Vitrinite Reflectance

Essential for the calculation of coke stability is the knowledge of the rank of coal, most popularly discussed in terms of reflectance (of vitrinite, the predominant maceral) Small-scale coking tests (by Schapiro and Gray) of mixtures of closely graded constituents (vitrinite of a narrow reflectance range) demonstrated that the strength of coke depends upon the rank of vitrinite, more precisely the reflectance distribution and composition balance index (inerts/reactives ratio).

The basic system has been revised and modified by others, some proposing a simplified prediction method in which the rank parameter is represented by mean maximum reflectance of vitrinite $R_c max$.

In Australian practice a distinction is made within the vitrinite group to accommodate differences in gelification and/or reflectance between the various members of that group.

Vitrinite-A consists of the higher reflecting and less gelified macerals telinite and telocollinite.

Vitrinite-B is highly gelified material, such as desmocollinite, and to a lesser extent, gellocollinite and corpocollinite.

Such/discrimination is not practised by all Petrographers and therefore considerable differences have been reported in ICCP exercises.

In a case study (commissioned by RCC) it was found that maceral composition considerable differ amongst the various particle sizes from over 19.0mm to below 0.5mm. These differences re particularly well displayed by the maceral groups Inertinite and Vitrinite which, from the coarsest to the finest size fraction reverse their respective percentages in the Bulli Seam.

The reason for the high proportion of inertinite in the coarse fraction is the toughest of dull ad durable (Durain) coal. In contrast, the vitrinite-rich bright portions of the coal (Vitrain) are brittle and thus concentrate in the fines. However, Vitrinite-B (Desmocollinite) does not contribute significantly to the increase in total vitrinite.

Since Vitrinite-A has a higher reflectance than Vitrinite-B, the reflectance values of A and B, measured in proportion to their respective frequencies, rise in the finer size fractions. This increase in the reflectance of Vitrinite-A in the finer size fractions is due to the relative smallness of Vitrinite-A in the coarse lumps, compounded by it being surrounded by colloidal democollinite. The latter has absorbed resinous bituminous substances which not only reduce it's own reflectance but also tend to affect the small inclusions of Vitrinite-A.

By contrast, the high proportion of Vitrinite-A in the finer sizes originates in bright coal (vitrain bands), concentration of telocollinite and talinite, relatively free of colloids which could lower their reflectance.

Thus, if sampling and/or sample preparation is biased (due to size segregation) a most unexpected distortion in rank and rank related properties can occur.

The more systematic differences in reflectance values are due to measurements of "random" or "average" reflectance with or without oil immersion. Such practices area all correct in their own right, however, they become anomalous if the full experimental details are lost or mis-translated.

CONCLUSION

The global trade in raw materials has accelerated the growth of industrial giants in the new-world. Increasing competition will demand higher grade raw materials which in turn will focus more critical attention on testing procedures.

The paper has reviewed some aspects of the traditionally well established although sometimes misunderstood procedures as to their intent, if not methodology. Important tests such as the Ropper Expansion test for the measurement of carbonisation pressure propensity has been omitted for considerations of time.

Wall pressure in coke ovens especially the extra tall ovens is of great concern. Case histories of the sometimes tortuous paths that coal exporters travel in order to demonstrate the technical integrity of their products occasionally identify commercially significant features hitherto unappreciated.

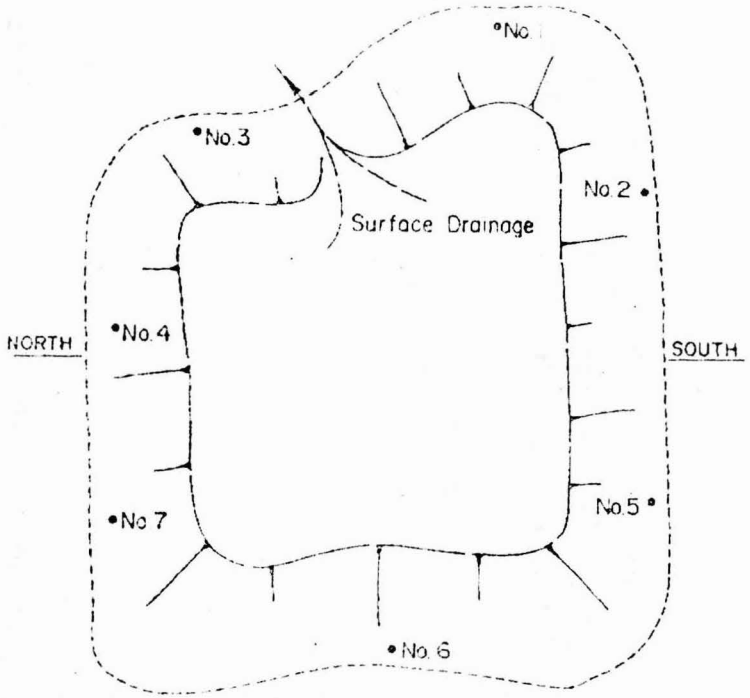
Such an unexpected bonus came about during blendability trials at a steel plant with very old coke ovens. High strength coke was sought with mandatory low reactivity and of course large lump size.

The blend formulated needed to be high in rank with the commensurate low volatile matter, which caused understandable concern with regards to possible wall damage. Pre-production trials to introduce a new coal into the existing blend demonstrated that not only had the new coal negligible carbonisation pressure but it also possessed considerable capacity to soak up the peak pressure of the original blend.

ACKNOWLEDGEMENT

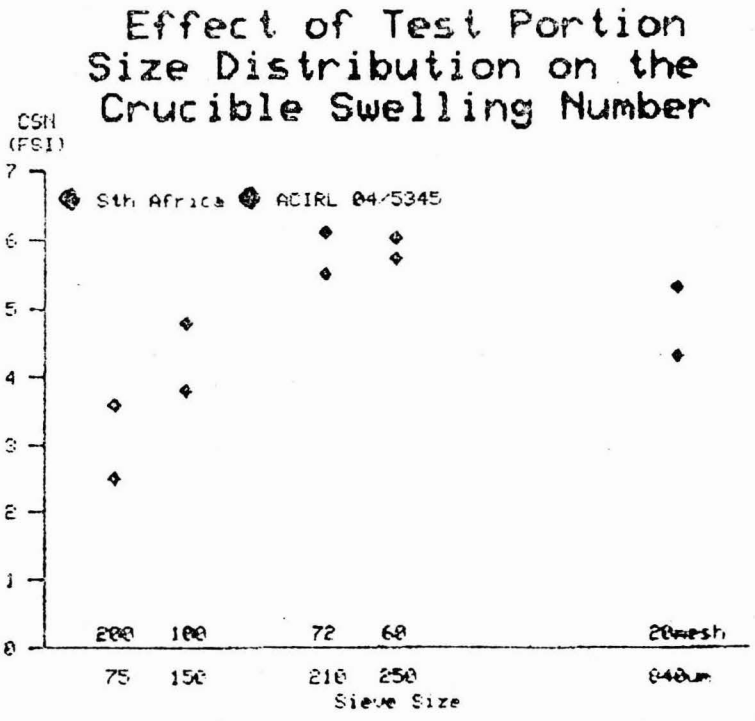
The data presented are extracts from Laboratory and Pilot scale carbonisation studies conducted by Internationally recognised Research Establishments on behalf of Kembla Coal and Coke Pty Limited, in the course of their Export Market Development Program.

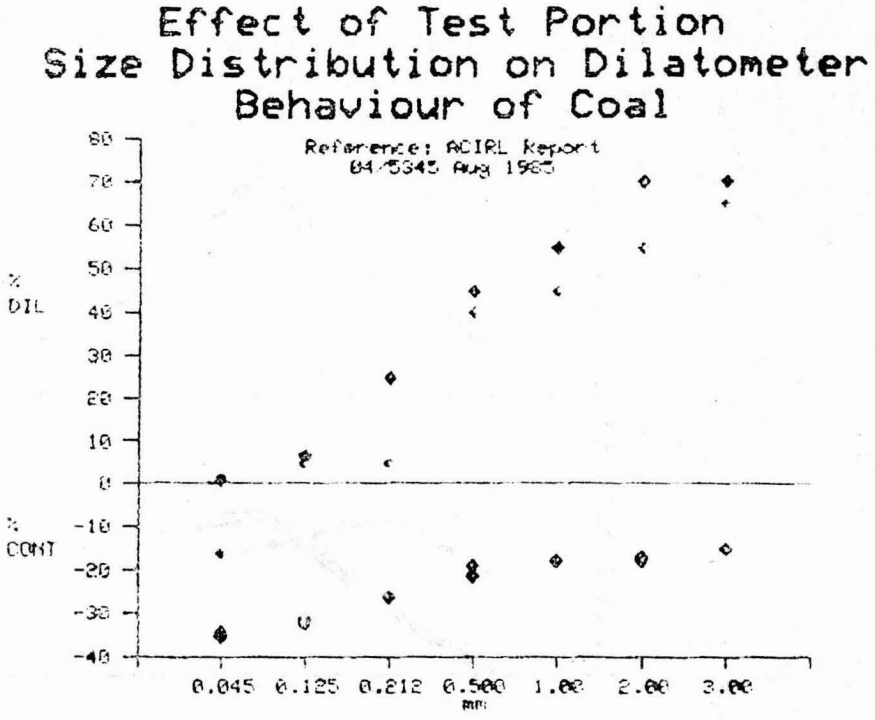
The views expressed are those of the author and not necessarily KCC's. Permission to publish is gratefully acknowledged with special thanks for funding of the numerous investigations and case studies to clarify anomalous interpretation of certain laboratory results.

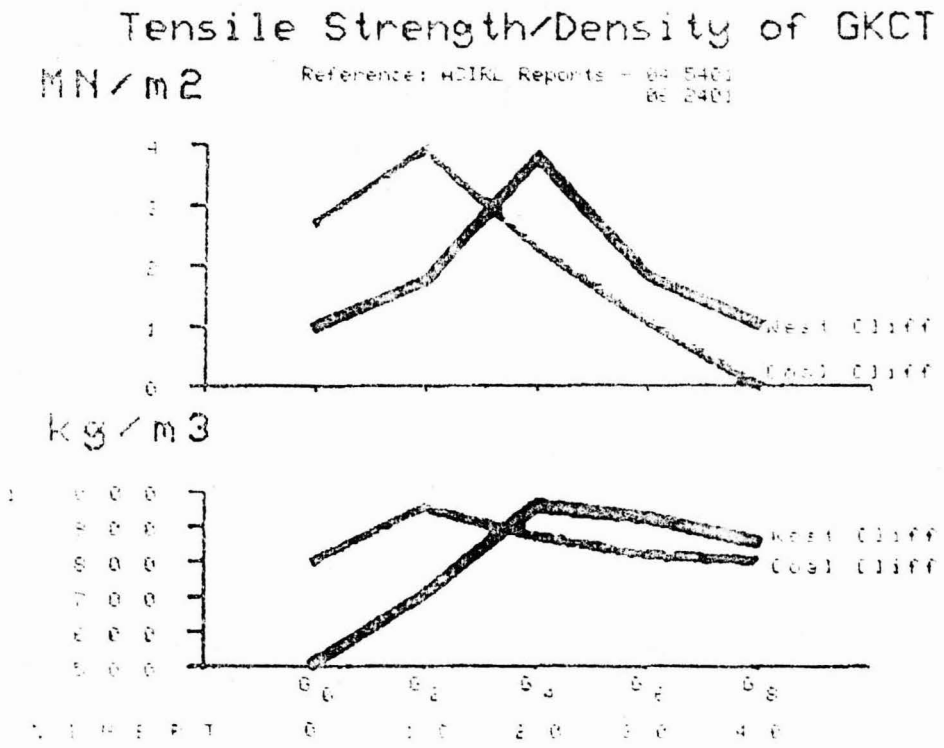


Not to scale

WEST CLIFF COLLIERY
No. 1 STOCKPILE
INITIAL SAMPLE POINTS


Effect of Test Portion Size Distribution on the Crucible Swelling Number
CSN (FSI)
7
6
5
4
3
2
1
0
Sth Africa
ADIRL 04/5345
200
100
72
60
20wash
75
150
210
250
840um
Sieve Size


Effect of Test Portion
Size Distribution on Dilatometer
Behaviour of Coal
Reference: ACIRL Report
% DIL
% CONT
80
70
60
50
40
30
20
10
0
-10
-20
-30
-40
0.045
0.125
0.212
0.500
1.00
2.00
3.00
mm


Tensile Strength/Density of GKCT
MN/m2
Reference: HDIRL Reports
West Cliff
Coal Cliff
kg/m3
West Cliff
Coal Cliff

Vitrinite Reflectance of Bulli Seam Coal - Sample No.9310

Ref: C F K Diessel, Report to KCC, May 1982

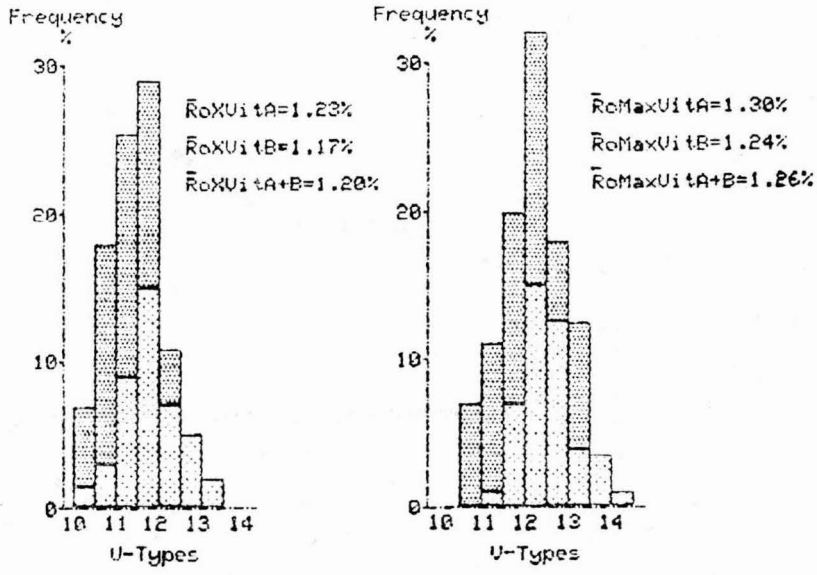


Figure 1: Vitrinite, Inertinite and Mineral Matter Distribution by Grain Size

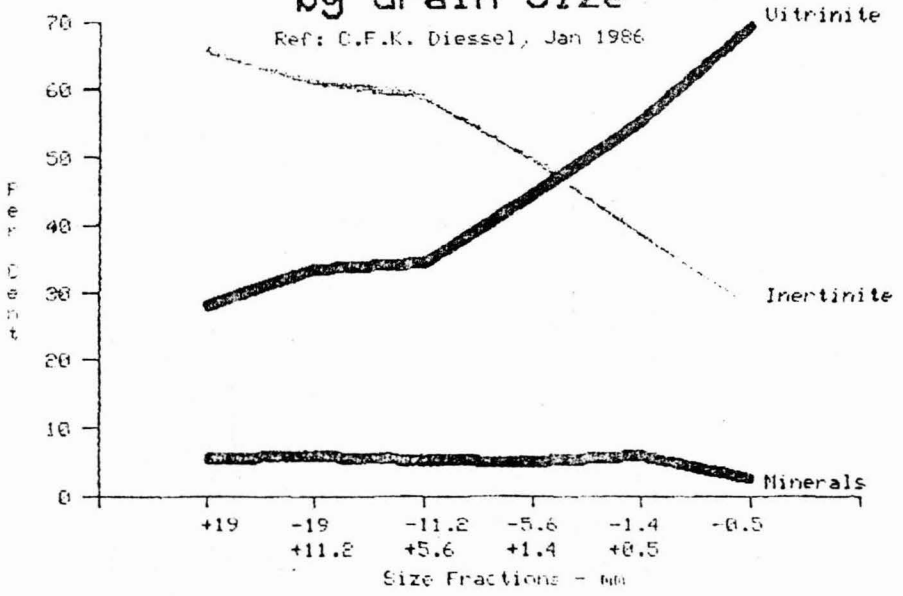


Figure 2: Vitrinite A and Vitrinite B Distribution by Grain Size

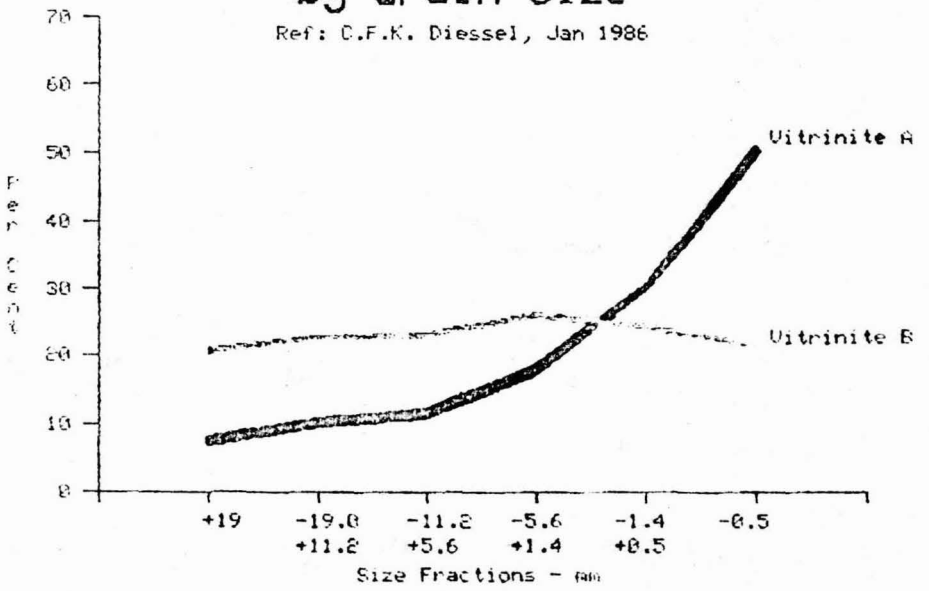
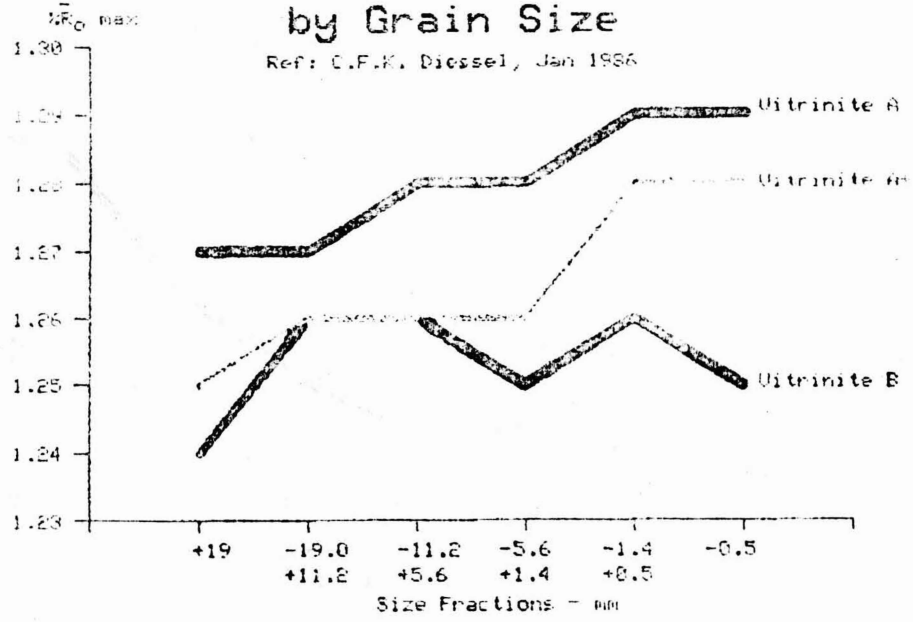


Figure 3: Reflectance of Vitrinite A and Vitrinite B Distribution by Grain Size



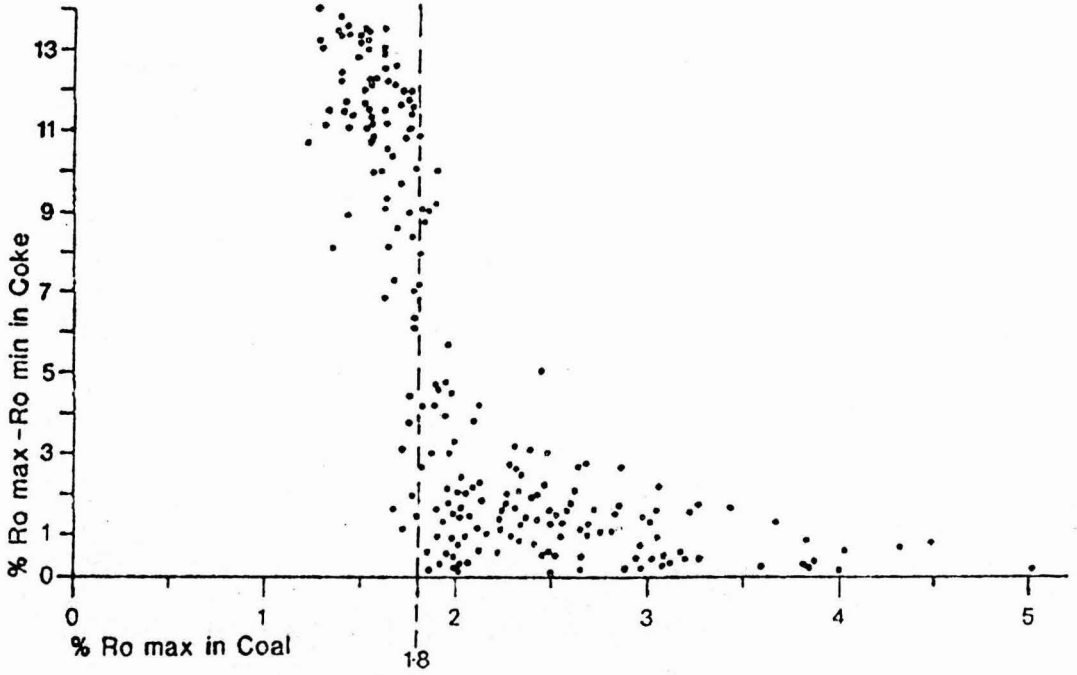


FIGURE 1